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(54) **Title of the invention:** Styrene thermoplastic resin composition and styrene thermoplastic molded product prepared by using the same

(57) **Summary**

Problem

To present a styrene thermoplastic resin composition that can give a molded product with excellent scratch resistance, wear resistance and bleeding resistance and, styrene thermoplastic molded product prepared by using the same.

Procedure for solution

Styrene thermoplastic resin composition consisting of the following (A) ~ (D) and a styrene thermoplastic molded product prepared by using the same. (A) 10 ~ 80 parts by weight of a graft copolymer obtained by graft polymerization of a mixture of an aromatic vinyl compound, a vinyl cyanide compound and a monomer that can polymerize with these, in presence of a rubbery polymer, (B) 20 ~ 90 parts by weight of a copolymer obtained by polymerizing a mixture of an aromatic vinyl compound, a vinyl cyanide compound and a monomer that can copolymerize with these. (Quantity of (A) and (B) together is 100 parts by weight), (C) silicone oil and (D) a compatibilizer consisting of modified polyolefin and / or a homopolymer and /or copolymer of a linear or alicyclic (metha) acrylic ester having at least 10 carbon atoms.

Scope of the invention

Claim 1

Styrene thermoplastic resin composition consisting of the following (A) ~ (D) and a styrene thermoplastic molded product prepared by using the same. (A) 10 ~ 80 parts by weight of a graft copolymer obtained by graft polymerization of a mixture of an aromatic vinyl compound, a vinyl cyanide compound and a monomer that can polymerize with these, in presence of a rubbery polymer, (B) 20 ~ 90 parts by weight of a copolymer obtained by polymerizing a mixture of an aromatic vinyl compound, a vinyl cyanide compound and a monomer that can copolymerize with these. (Quantity of (A) and (B) together is 100 parts by weight), (C) silicone oil and (D) a compatibilizer consisting of modified polyolefin and /or a homopolymer and/or copolymer of a linear or alicyclic (metha) acrylic ester having at least 10 carbon atoms.

Claim 2

Styrene thermoplastic resin composition mentioned in claim 1 above comprising 0.1 ~5 parts by weight of silicone oil (C) and 0.01 ~0.5 parts by weight of (D) a compatibilizer made from a modified polyolefin and/or a homopolymer and/or copolymer of a linear or alicyclic (metha) acrylate ester having more than 10 carbon atoms, as against 100 parts by weight of graft copolymer (A) and copolymer (B) together.

Claim 3

Styrene thermoplastic resin composition mentioned in claims 1 or 2 above, where the modified polyolefin in the compatibilizer (D) is a polyethylene chloride with degree of chlorination of 20 ~ 50%.

Claim 4

Styrene thermoplastic resin composition mentioned in claims 1 or 2 above, in which the modified polyolefin in compatibilizer (D) is acid-modified polyethylene wax with its extent of acid modification in the range 0.5 ~ 30 KOH mg/g.

Claim 5

Styrene thermoplastic resin composition mentioned in claims 1 or 2 above, in which the modified polyolefin in compatibilizer (D) is styrene-ethylenebutene-styrene block polymer and /or styrene-butadiene-styrene block polymer with styrene content of 10 ~ 80 weight %.

Claim 6

Styrene thermoplastic resin composition mentioned in claim 1 or 2 above, in which the linear or alicyclic (metha) acrylate ester and its copolymer in the compatibilizer (D) is a tricyclodecane methacrylate / methyl methacrylate copolymer with copolymerization ratio of 2/8 ~ 4/6.

Claim 7

Styrene thermoplastic resin composition mentioned in any of the claims 1 ~ 6 above, in which acrylic rubber is used as the rubbery polymer.

Claim 8

Styrene thermoplastic resin molded product using styrene thermoplastic resin composition mentioned in any of the claims 1 ~ 7 mentioned above.

Detailed description of the invention

[0001]

Technical fields belonging to this invention

This invention is concerned with styrene thermoplastic resin composition and molded prepared from this composition.

[0002] Conventional techniques

Conventionally the method of addition of silicone oil to styrene thermoplastic resin is adopted widely for improving the scratch resistance and wear resistance. If, however, more silicone oil than a certain quantity is added to improve the scratch resistance further, the molded product bleeds silicone oil from the surface. In other words, the silicone oil present on the surface condenses and becomes visible.

[0003] The problems this invention sought to solve

When silicone oil is added to improve scratch resistance, there was a problem of bleeding silicone oil from the surface of the molded product and this affected the external appearance of the product. The aim of this invention is to present a styrene thermoplastic resin that maintains scratch resistance and wear resistance without bleeding silicone oil.

[0004] Procedure for solving the problems

This invention is concerned with styrene thermoplastic resin composition and molded product made from this composition. Styrene thermoplastic resin composition consisting of the following (A) ~ (D) and a styrene thermoplastic molded product prepared by using the same. (A) 10 ~ 80 parts by weight of a graft copolymer obtained by graft polymerization of a mixture of an aromatic vinyl compound, a vinyl cyanide compound and a monomer that can polymerize with these, in presence of a rubbery polymer, (B) 20 ~ 90 parts by weight of a copolymer obtained by polymerizing a mixture of an aromatic vinyl compound, a vinyl cyanide compound and a monomer that can copolymerize with these. (Quantity of (A) and (B) together is 100 parts by weight), (C) silicone oil

and (D) a compatibilizer consisting of modified polyolefin and /or a homopolymer and/or copolymer of a linear or alicyclic (metha) acrylic ester having at least 10 carbon atoms.

[0005] In case of this invention, the resin composition contains 0.1 ~5 parts by weight of silicone oil (C) and 0.01 ~0.5 parts by weight of (D) a compatibilizer made from a modified polyolefin and/or a homopolymer and/or copolymer of a linear or alicyclic (metha) acrylate ester as against 100 parts by weight of graft copolymer (A) and copolymer (B) together.

[0006] In this invention, the modified polyolefin in the compatibilizer (D) is a polyethylene chloride with degree of chlorination of 20 ~ 50%.

[0007] In case of this invention, the modified polyolefin in the compatibilizer (D) is acid-modified polyethylene wax with its extent of acid modification in the range 0.5 ~ 30 KOH mg/g.

[0008] In case of this invention, it is better that the modified polyolefin in compatibilizer (D) is styrene-ethylenebutene-styrene block copolymer and /or styrene-butadiene-styrene block copolymer with styrene content of 10 ~ 80 weight %.

[0009] In case of this invention, the linear or alicyclic (metha) acrylate ester and its copolymer with more than 10 carbon atoms in the compatibilizer (D) is a tricyclodecane methacrylate / methyl methacrylate copolymer with copolymerization ratio of 2/8 ~ 4/6.

[0010] In this invention, it is better to use acrylic rubber as the rubbery polymer.

[0011] **Implementation of this invention**

Styrene thermoplastic resin composition and molded product made from such composition are explained below in detail. Examples of the graft polymer (A) are acrylonitrile-acrylic rubber-styrene copolymer (AAS resin), acrylonitrile-ethylene propylene rubber-styrene graft copolymer (AES resin), acrylonitrile-butadiene-styrene (ABS resin). Here acrylic rubber, ethylene propylene rubber, butadiene rubber are the examples of the rubbery polymer. Out of these, acrylic rubber is preferable. Especially AAS resin obtained by graft polymerization of acrylonitrile and styrene on acrylic rubber is still better as far as weather resistance is concerned.

[0012] α -substituted styrene like α -methyl styrene, α -ethyl styrene; nuclear substituted styrene like chlorostyrene, vinyl styrene, t-butyl styrene can be used as aromatic vinyl compounds of copolymer (B) of this invention. Further, acrylonitrile, methacrylonitrile can be used as the vinyl cyanide compounds.

[0013] Examples of silicone oil (C) are dimethyl silicone oil, fluorine modified silicone oil, amino modified silicone oil and epoxy silicone oil. However silicone oil with viscosity 1000 ~ 100,000 cSt is desirable. That with viscosity 1000 ~ 30000 cSt is more desirable. If the viscosity is less than 1000 cSt, there is a tendency of lesser improvement in scratch resistance. If, however, the viscosity exceeds 100,000 cSt, there is a tendency of bleeding.

[0014] Modified polyolefin and / or homopolymer and/or copolymer of linear or alicyclic (metha)acrylate ester (D) is used for improving the compatibility between graft copolymer (A)/copolymer (B) and silicone oil (C).

[0015] Its specific examples are polyethylene with 20 ~ 50 % chlorination, acid modified polyethylene wax with acid modification of 0.5 ~ 30 KOH mg/g. Styrene-ethylenebutene-styrene

block polymer with 10 ~ 80 weight % styrene content and/ or styrene-butadiene-styrene block polymer tricyclodecane methacrylate copolymer with ratio of polymerization in the range 2/8 ~ 4/6.

[0016] In the above, if the chlorination of chlorinated polyethylene is less than 20 %, the compatibility of graft copolymer (A)/copolymer (B) and chlorinated polyethylene reduces and it cannot function as compatibilizing agent for graft copolymer (A)/copolymer (B) and silicone oil (C) and there is a tendency to bleed silicone oil. Further, if the chlorination exceeds 50 %, the compatibility between polyethylene and silicone oil (C) reduces and there is a tendency to bleed silicone oil.

[0017] If the acid modification of acid modified polyethylene wax is less than 0.5 KOH mg/g, the compatibility of graft copolymer (A)/copolymer (B) and acid modified polyethylene wax reduces and it cannot function as compatibility agent for graft copolymer (A)/copolymer (B) and silicone oil (C) and there is a tendency to bleed silicone oil. Further, if the acid modification exceeds 30 KOH mg/g, the compatibility of acid modified polyethylene wax and silicone oil (C) reduces and similar bleeding of silicone oil may occur.

[0018] If the styrene content of styrene-ethylene butene-styrene block polymer and /or styrene-butadiene-styrene block copolymer is less than 10 weight %, the compatibility of graft copolymer (A)/copolymer (B) and styrene-ethylenebutene-styrene block polymer and /or styrene-butadiene-styrene block polymer reduces and it cannot function as compatibility agent for graft copolymer (A)/copolymer (B) and silicone oil (C) and there is a tendency to bleed silicone oil. If, however, this quantity exceeds 80 weight %, the compatibility of graft copolymer (A)/copolymer (B) and styrene-ethylene butene-styrene block polymer and /or styrene-butadiene-styrene block polymer reduces and there is a tendency to bleed silicone oil.

[0019] If the copolymerization ratio of tricyclodecanemethacrylate/methyl methacrylate copolymer is less than 2/8, the compatibility of tricyclodecanemethacrylate /methylnmethacrylate copolymer and silicone oil reduces and it may not function as compatibility agent for graft copolymer (A)/copolymer (B) and silicone oil (C), and there is a tendency to bleed silicone oil. If this value exceeds 4/6, the compatibility of tricyclodecanemethacrylate /methylnmethacrylate copolymer reduces and there is a tendency to bleed silicone oil.

[0020] Content of graft copolymer (A) of this invention is 10 ~ 80 parts by weight per 100 parts by weight of total of graft copolymer (A) and copolymer (B). Desirable value is 15 ~ 60 parts by weight and more desirable value is 20 ~ 50 parts by weight. If the content of graft copolymer (A) is less than 10 parts by weight, the impact resistance reduces and if it exceeds 80 parts by weight, the fluidity and molding properties are affected.

[0021] Content of copolymer (B) is such that it is 20 ~ 90 parts by weight per 100 parts by weight of total of graft copolymer (A) and copolymer (B). Desirable value is 40 ~ 85 parts by weight and more desirable value is 50 ~ 80 parts by weight. If the quantity of copolymer (B) is less than 20 parts by weight, the molding properties are affected and if it exceeds 90 parts by weight, the impact strength is affected.

[0022] Quantity of silicone oil (C) in this invention is 0.1 ~ 5 parts by weight per 100 parts by weight of total of graft polymer (A) and copolymer (B). Desirable value is 0.3 ~ 3 parts by weight and more desirable value is 0.5 ~ 1.5 parts by weight. If the quantity of silicone oil is less than 0.1 parts by weight, the scratch resistance reduces and if it exceeds 5 parts by weight, there is a tendency of bleeding of silicone oil.

[0023] Desirable proportion of compatibility agent (D) is 0.01 ~ 0.5 parts by weight. More desirable value is 0.03 ~ 0.3 parts by weight and highly desirable value is 0.05 ~ 0.15 parts by weight. If this is less than 0.01 parts by weight, there is a tendency of bleeding silicone oil and if it exceeds 0.5 parts by weight, the scratch resistance reduces.

[0024] In addition to the necessary components (A) ~ (D) of the resin composition of this invention, additives like other resins, coloring agents, phenol antioxidants, smoothening agents, ultra violet absorbers can be added.

[0025] By making use of the resin composition of this invention, styrene thermoplastic moldings like automobile related parts, eaves gutters or building parts can be prepared.

[0026]

Application examples

This invention is concretely explained by giving application examples but it is not restricted to these examples. Further, in the following discussion, 'part' means 'part by weight' and '%' means 'weight %'. The abbreviations of compounds used in the application examples are as follows.

St:Styrene

BuA:Butyl acrylate

AN: Acrylonitrile

MMA: Methyl methacrylate

HDA: 1,6-hexane dioldiacrylate

AMA: Acrylmethacrylate

TDM: t-dodecylmercaptan

KPS: Potassium persulphate

DAS: di-2-ethylhexylsulpho sodium succinate

[0027] Application example 1

Preparation of graft polymer (A)

Sampled polymerization reaction mixture was dried by using an infrared moisture meter, and the weight of non-volatile matter was measured. The degree of polymerization was determined from this value and the initial proportion by using the following formula.

$$\text{Degree of polymerization of } n \text{ layer} = ((\alpha \times \beta \div \gamma - \delta) / \epsilon) \times 100 \%$$

Where

n: integer from 1 to 5

α : Total weight of the reaction mixture existing in the polymer system at the time of completion of n th stage polymerization

β : Weight of non-volatile matter from the reaction mixture taken out at the time of completion of polymerization of n th stage

γ : Weight of the reaction mixture taken out at the time of completion of polymerization of n th stage

δ : Total weight of non-volatile matter existing in the polymer system at the time of completion of polymerization up to (n-1) th stage

ϵ : Total of the monomer and multifunctional monomer added freshly at the n th stage and that of unreacted monomer mixture existing in the polymer system at the time of completion of (n-1) th stage

Further, when n – 1 is 0 in the above case, the value of δ is 0 and ϵ is the total quantity of monomer added in the first stage.

[0028] Stage 1

1614 g distilled water and 35 g DAS were taken in a 4-liter flask equipped with a reflux condenser and the temperature was raised to 40°C while stirring at 320 rpm. Nitrogen gas was passed for about one hour through the water, while maintaining the temperature at 40°C, for replacing the dissolved oxygen from the water. After this the temperature was raised to 70°C under a current of nitrogen and 25.3 g of 1.19 weight % aqueous KPS solution was added. This was followed by the addition of a mixture of 371.03 g St and 0.565 g HDA at the rate of 9.3 g/min. After the addition was complete, it was held at 70°C for 2 hours. The percent polymerization at this stage was 96.5 %.

Stage 2

Then 25.3 g of 1.1 weight % aqueous solution of KPS was added and a mixture consisting of 371.03 g BuA and 0.458 g HDA was added at the rate of 9.3 g /min. Then it was held at 70°C for 2.5 hours. The overall degree of polymerization of monomer mixture in the second stage and unreacted portion in the monomer mixture from the first stage was 96.2 %. The degree of polymerization of total monomer used in the first stage and second stage was 98.0.

Stage 3

Then 25.8 g of 2.9 weight % aqueous solution of KPS and 1.0 g DAS were added and then a mixture consisting of 68.7 g St, 13.8 g BuA and 1.736 g HDA was added at the rate of 9.3 g/min and it was held at 70°C for 4 hours. The degree of polymerization of unreacted monomer at the end of the second stage and newly added monomer mixture in the third stage together (this is known as 'degree of polymerization in stage 3') was 98.8.

Stage 4

Then 25.4 g of 1.4 weight % aqueous solution of KPS was added and a mixture consisting of 112.1 g St, 35.4 g AN and 4.399 g AMA was added at the rate 9.3 g /min and it was held at 70°C for 1.5 hour. Degree of polymerization of unreacted monomer at the end of stage 3 and monomer mixture

newly added in stage 4 together (this is known as 'degree of polymerization in stage 4') was 93.0 % and the degree of polymerization of monomer from stage 1 to stage 4 was 99.1 %.

Stage 5

Then 25.3 g of 1.3 weight % aqueous solution of KPS was added and a previously prepared solution was added at the rate of 9.3 g/min. This previously prepared mixture was prepared by adding 51.1 g MMA, 194.2 g St, 95.4 g of AN and 2.308 g of TDM to 250 g distilled water and then adding 1.908 g of DAS and stirring it sufficiently to form an emulsion. This mixture was added at the rate of 13 g/min and it was held at 70°C for 1.5 hour. Degree of polymerization of unreacted monomer at the end of stage 4 and monomer added in stage 5 together (this is known as 'degree of polymerization in stage 5') was 95.5 %. Then the entire latex thus obtained was slowly added to 3000 g of 1.3 weight % aqueous aluminium sulphate solution at 80°C while stirring vigorously, during a period of 30 minutes. The separated matter was dried by removing water, to get styrene powder with multilayered structure.

[0029] Preparation of copolymer B

1500 parts of ion exchanged water, 30 parts of 10 weight % aqueous solution of calcium phosphate and 0.4 parts of 12.5 weight % aqueous solution of dodecylbenzene sulphonic acid sodium salt were taken in a 4-liter reaction vessel and mixed by stirring. Then 855 parts styrene, 360 parts acrylonitrile, 285 parts methyl methacrylate, 3.6 parts of t-dodecylmercaptan, 7.5 parts dilauryl peroxide and 1.1-bis (t-butyl peroxy) 3,3,5-trimethylcyclohexane were mixed and dissolved and then added. After substitution by nitrogen, it was subjected to suspension polymerization at 65°C for 8 hours and then the temperature was raised to 110°C. After conducting polymerization for 2 hours, it was cooled. This suspension was dried to remove water, to get polymer (B).

[0030] 1.0 part of silicone oil (C), 0.05 part of chlorinated polyethylene with degree of chlorination of 30% (D), 0.3 part of antioxidant (Kitsutomi Fine Chemicals, Tominox SSR), 0.3 part of photo stabilizer (Kitsutomi Fine Chemicals, Tomisorb77), 2.0 parts of smoothening agent (Kao Co., WAX-EB), 6.0 parts of coloring agent (Carbon black master batch with carbon content of 35 weight %, Trade name) were compounded with 37 parts of the graft copolymer (A) obtained as above and 63 parts of copolymer (B) and palletized by conducting fused kneading by biaxial extrusion (Manufactured by Ikegu Tekkou: PCM-30). These pellets were subjected to injection molding and test pieces were prepared for measurement. The properties were measured by the following methods. The results are presented in Table 1.

[0031] (1) Strength against impact (J/m)

Izot impact strength of 1/8 inch test piece (with notch) was measured according to JIS K7110

(2) Bending strength (Mpa)

According to JIS K7203.

(3) Fluidity (g/ 10 min)

Flow in 10 minutes, under conditions of 220°C, 10 kg load was measured according to JIS K7210.

(4) Coefficient static friction

A polyethylene foam sheet of thickness 1 mm was pasted to a sheet to make a reference surface. A flat test piece of 90x90x3 t was prepared from the thermoplastic resin composition. This was placed on the reference surface and 1 kg load was added to the test piece. The reference surface was slowly inclined with the test piece placed on it and the angle θ at which the test piece starts to move was noted. Coefficient of static friction was expressed as $\tan \theta$.

(5) Bleeding property

A flat test piece of 90x150x 3t was prepared by a pressing cycle of 120 seconds, with resin temperature of 230°C, by means of a pressing machine (Nippon Seikosho: J75SA). The pressing frequency was counted and pressing frequency for bleeding of silicone oil was measured.

[0032] Application examples 2 ~ 10

Graft copolymer (A) and copolymer (B) obtained as in application example 1, were used. After blending with silicone oil (C), compatibility agent (D), antioxidant, photo-stabilizer, smoothening agent and coloring agent as shown in Table 2, biaxial fused extrusion was carried out (Ikekai Tekkou:PCM-30), pellets were obtained. Test pieces were prepared from these pellets by injection molding, and the evaluation was carried out as above. The results are presented in Table 1 and 2.

[0033] Comparison examples 1 ~ 7

Graft copolymer (A) and copolymer (B) obtained as in application example 1, were used. After blending with silicone oil (C), compatibility agent (D), antioxidant, photo-stabilizer, smoothening agent and coloring agent as shown in Table 3, biaxial fused extrusion was carried out (Ikekai Tekkou:PCM-30), pellets were obtained. Test pieces were prepared from these pellets by injection molding, and the evaluation was carried out as above. The results are presented in Table 3.

[0034]

Table 1

| Polymer composition | Unit | Appl. | Appl. | Appl. | Appl. | Appl. | Appl. |
|---------------------|-------|-------|-------|-------|-------|-------|-------|
| | | Ex. 1 | Ex. 2 | Ex. 3 | Ex. 4 | Ex. 5 | Ex. 6 |
| Graft polymer (A) | Parts | | | | | | |

| | | | | | | | |
|--------------------------------|---------|--|--|--|--|--|--|
| Copolymer (B) | | | | | | | |
| Silicone oil (C) | | | | | | | |
| Compatibility agent (D) | | | | | | | |
| Antioxidant | | | | | | | |
| Photo-stabilizer | | | | | | | |
| Smoothing agent | | | | | | | |
| Coloring agent | | | | | | | |
| Impact strength | J / m | | | | | | |
| Bending strength | Mpa | | | | | | |
| Fluidity | G/10min | | | | | | |
| Coefficient of static friction | - | | | | | | |
| Bleeding | times | | | | | | |

[0035]

Table 2

| Polymer composition | Unit | Appl. Ex. 7 | Appl. Ex. 8 | Appl. Ex. 9 | Appl. Ex. 10 |
|-------------------------|-------|-------------|-------------|-------------|--------------|
| Graft polymer (A) | Parts | | | | |
| Copolymer (B) | | | | | |
| Silicone oil (C) | | | | | |
| Compatibility agent (D) | | | | | |
| Antioxidant | | | | | |
| Photo-stabilizer | | | | | |
| Smoothing agent | | | | | |
| Coloring agent | | | | | |
| Impact strength | J / m | | | | |
| Bending strength | Mpa | | | | |

| | | | | | |
|--------------------------------|---------|--|--|--|--|
| Fluidity | G/10min | | | | |
| Coefficient of static friction | - | | | | |
| Bleeding | times | | | | |

[0036]

Table 3

| Polymer composition | Unit | Comp. Ex. 1 | Comp. Ex. 2 | Comp. Ex. 3 | Comp. Ex. 4 | Comp. Ex. 5 | Comp. Ex. 6 | Comp. Ex. 7 |
|--------------------------------|---------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|
| Graft polymer (A) | Parts | | | | | | | |
| Copolymer (B) | | | | | | | | |
| Silicone oil (C) | | | | | | | | |
| Compatibility agent (D) | | | | | | | | |
| Antioxidant | | | | | | | | |
| Photo-stabilizer | | | | | | | | |
| Smoothening agent | | | | | | | | |
| Coloring agent | | | | | | | | |
| Impact strength | J / m | | | | | | | |
| Bending strength | Mpa | | | | | | | |
| Fluidity | G/10min | | | | | | | |
| Coefficient of static friction | - | | | | | | | |
| Bleeding | times | | | | | | | |

[0037]

- * Silicone oil: Toray Dow Corning Silicone : SH-200 (Viscosity 10000 cSt)
- * Compatibility agent (D): *1 Chlorinated polyethylene with degree of chlorination of 30%
*2 Oxygen modified polyethylene wax of oxygen modification

of 10 KOH mg/g

*3 Tricyclodecanemethacrylate / methyl methacrylate

copolymer of degree of polymerization = 3/7

*4 Styrene-ethylenebutene-styrene block copolymer of 10 %

styrene content

*5 Oxygen modified polyethylene wax with oxygen

modification = 0 KOH mg/g

- * Antioxidant: Kitsutomi Fine Chemicals: Tominox SSR
- * Photo-stabilizer: Kitsutomi Fine Chemicals: Tomisorb 77
- * Smoothing agent: Kao Co. :WAX-EB
- * Coloring agent: Carbon black master batch (Carbon content 35 weight %)
- * Bleeding property: (-) in the Table means that bleeding does not occur even after pressing for 50 times.

[0038] Results of this invention

Thermoplastic resin composition of this invention has excellent scratch resistance and therefore it is difficult to get scratches on products made from it during handling and use, and this maintains its external appearance. It can thus provide material that can be used in automobile parts, eaves gutters, material for housing and so on.